III.F Manufacturing

III.F.1 Tape-Calendering Manufacturing Processes for Multi-Layer Thin-Film Solid Oxide Fuel Cells

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Objectives

- Develop a manufacturing process along with an advanced cell configuration that will contribute to significantly lowering cell first cost while improving cell robustness, life, reliability, and maintainability

Approach

- Examine and optimize cell fabrication process based on tape calendering
- Map cell fabrication process
- Identify critical process parameters for yield, performance, etc.
- Examine effects of cell footprint scale-up
- Modify fabrication parameters to produce multi-layer cells and unitized cells for characterization
- Assess the properties of the fabricated cells using modeling and electrochemical testing for screening
- Examine destructive and nondestructive testing requirements for various manufacturing process steps
- Develop unitized cells and demonstrate electrochemical performance under specified conditions of temperature, gas flow rate, utilization, and current density
- Model and validate flow field designs using computational fluid dynamics (CFD), electrochemical and physical testing
- Examine issues of fabricating stamped metallic interconnect sheets
  - Establish design limits
  - Evaluate manufacturing process

Accomplishments

- Cathode and anode improvements, such as modifications to powder morphologies and compositions, processing improvements and formulation changes, have led to significantly improved solid oxide fuel cell (SOFC) performance, especially in the 650 to 700°C range, for anode-supported cells manufactured by tape calendering
  - Cell power density was increased from 0.243 W/cm² to 0.892 W/cm² for 650°C operation
  - Cells were operated at greater than 70% fuel utilization

- Cells were fabricated that exhibited a 3x improvement in flexure strength while maintaining a peak power density of 0.914 W/cm²
- Flow field designs for unitized cells were analyzed and optimized for improved flow uniformity across the active area
  – 70% fuel utilization at a power density of greater than 0.2 W/cm² was demonstrated on a unitized cell operating on dilute hydrogen (64%)
- Several nondestructive evaluation techniques for flaw detection in multi-layer cells were evaluated; digital radiography was selected for further evaluation
- Developed forming models and investigated advanced joining methods, such as laser, electron-beam, and micro-TIG welding, for fabricating complex interconnect structures

Future Directions

- Work is completed on this project; appropriate process improvements will be incorporated in the work performed under the Solid State Energy Conversion Alliance SOFC Program (Cooperative Agreement DE-FC26-01NT41245)

Introduction

The overall objective of this project is to develop a low-cost tape-calendering process for manufacturing high-performance SOFCs using the unitized cell design. The innovative unitized cell concept has many attractive features, including simple stacking processes, ease of cell handling, improved maintainability, and high reliability. The tape-calendering process for cell manufacture has many desirable characteristics for low-cost and high-volume production: robustness, simplicity, scalability, automation, and simple quality control. For SOFC systems, the tape-calendering manufacturing process along with the unitized cell configuration will contribute to significantly lowering cell first cost while improving cell robustness, life, reliability, availability, and maintainability.

Approach

Two key aspects in developing a low-cost, high-performance SOFC technology are cell and interconnect fabrication. Any SOFC technology requires appropriate multi-layer ceramic fabrication and assembly methods to incorporate cell materials into a desired cell configuration. Appropriate methods are also required for making suitable interconnect structures that can be used to build a stack. The fabrication and assembly processes must ensure that no condition or environment in any process step destroys desired material characteristics of any of the components. Fabrication and assembly methodologies must attain the desired structural integrity, shape, electrical conductivity, and electrochemical performance of single cells and interconnect structures in the stack.

The overall approach of this project integrates two key elements to achieve the program objective: (a) the development of a process based on tape calendering for manufacturing multi-layer fuel cell components and (b) an innovative unitized cell design concept. The manufacturing approach is based on tape calendering as a core technology for fabricating thin-electrolyte, anode-supported cells. Tape calendering is the formation of a continuous sheet of tape of controlled size by squeezing of a softened thermoplastic material between two rolls. Components can be fabricated by calendering to contain multifunctional multilayers to enhance cell performance. The cell configuration is based on a unitized cell concept in which an individual cell is contained within a metallic housing with its own gas channels and manifolds to form a complete cell package. The fabrication process and cell configuration have all the characteristics required for low-cost production of high-performance cell packages that can be easily used for building stacks.

Results

Cathode and anode improvements, such as modifications to powder morphologies and compositions, processing improvements and

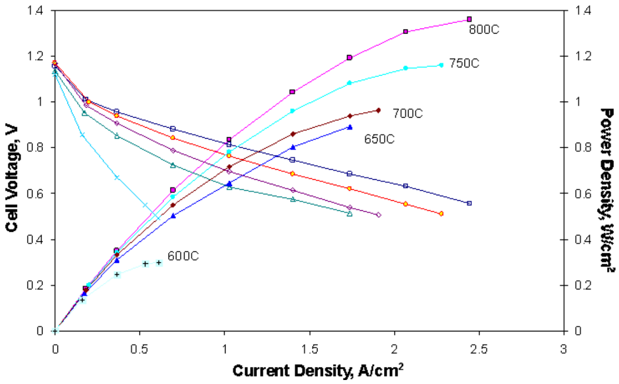

Figure 1. Polarization Curve from a Cell with an Improved Cathode; Pure hydrogen fuel with a fixed fuel flow rate of 67 cc/min and non-flowing air as oxidant

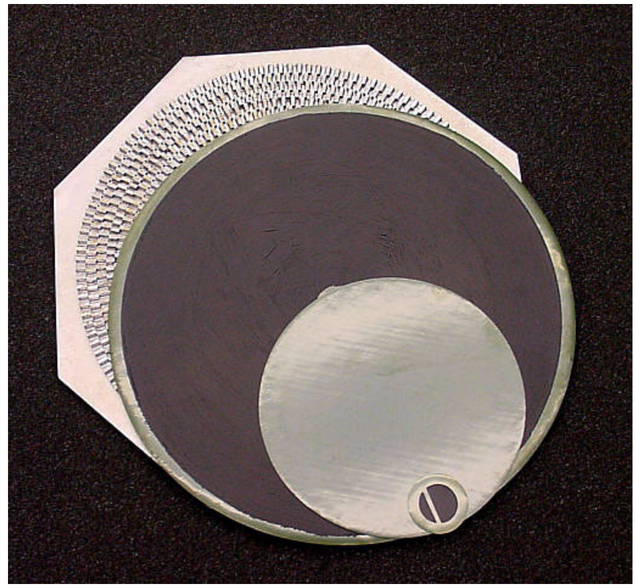

Figure 2. Photograph of a Tape-Calendered 8" Cell and Other Samples

formulation changes, and cell configurations have lead to significantly improved SOFC performance, especially in the 650 to 700°C range, for anode-supported cells made by calendering. By modifying the particle size distribution of the cathode constituents and then optimizing the firing temperature, cells were fabricated that showed increased performance from 0.250 W/cm² to nearly 0.9 W/cm² for 650°C operation. This performance was world-leading at the time of its reporting. A polarization curve from a cell of this design is presented in Figure 1. Anode advancements resulted in cell designs that exhibited stable performance at increased fuel utilization; fuel utilization increased from 75% to greater than 80% while the cell performance nearly doubled (increased from 380 to 650 mW/cm²). These improvements were focused around improvement of the porosity in the anode electrode by engineering the pore-forming additives in the individual layers for size, shape, amount, and distribution. Reactant flow to the electrolyte was improved. To manage the possible loss in mechanical strength from pore-former engineering, strength-improving approaches were investigated. Again, through engineering the individual layers, a 3x improvement in flexure strength was demonstrated with only minimal loss in electrochemical performance.

The tape-calendering cell fabrication process was examined and optimized. Detailed process maps were developed, and process parameters critical for yield, performance, etc. were identified. Fabrication of large-area cells was also attempted. Figure 2 shows as an example a photograph of an 8" cell made by calendering (along with an interconnect, a smaller bilayer, and a 1" button cell). The effects and issues related to cell footprint scale-up were explored.

Unitized cells were designed and tested to address the stacking issues of practical fuel cells. A significant number of cells were fabricated and tested to probe the many issues of stacking, including sealing, contact resistance, and flow distribution. Flow paths were designed that exhibited stable performance at high fuel utilizations (>70%) in electrochemical tests yet are expected to be formable with traditional stamping processes. Improved assembly processes led to unitized cell tests with maximum power densities of 0.3 W/cm² at fuel utilizations of 50%. This is nearly a 100% improvement in power density over the duration of the project.

In the area of non-destructive evaluation, several non-destructive imaging modalities, including infrared imaging, ultrasonic imaging, and digital radiography, were evaluated for their capabilities in defect recognition in SOFC materials in various stages of manufacture. Using radiography, a number of SOFC cells were followed through sequential stages of manufacture by tape calendering, from the bilayer tapes all the way to the fired cathodes on the bilayer substrate. Many different inhomogeneities were identified, characterized and cataloged. The short acquisition time and high image fidelity showed that digital radiography was very well suited to screening, cataloging, and monitoring surface and structural defects that occur throughout the manufacturing phases of SOFC components. The level of detail has shown to be sufficient to identify defects and density inhomogeneities at the level of 200 microns in a tape or cell. Furthermore, the digital data enables fast quantitative estimates of material thickness and scales of the inhomegeneities.

Potential processes for interconnect fabrication were assessed. Analytical and experimental means were established to determine these manufacturing process capabilities, requirements and gaps. Data were generated in the following areas.

- Formability of metallic interconnects and current collectors
- Flatness of metallic interconnects and current collectors
- Methods to assemble metallic interconnects, cell support sheet

Detailed process capability assessments were conducted using the following approaches: (a) Paper studies: survey of industrial mass production lines—consumer appliances and high-performance heat exchangers; consultation with industrial experts;
(b) Analytical: stack- and component-level stress analyses; finite element simulations;
(c) Experimental: component-level fabrication and testing; fabrication of prototype interconnect components; assembly of prototype interconnect components; testing of interconnect components.

In addition to theoretical and experimental determination of process capability, a sheet metal interconnect was manufactured by manufacturing methods similar to those intended for use in a mass production environment. The interconnect pan was stamped, then assembled by laser welding. A glass-sealed tape-calendered cell was tested for performance using this interconnect.

Conclusions

- Improved cathode and anode compositions and processes for producing cells by tape calendering were identified and validated.
- Cell strength was characterized and improved.
- Non-destructive evaluation techniques for tape-calendered cells were examined and identified for further investigation.
- Unitized cells were designed and tested; these cells achieved high fuel utilizations while maintaining other performance characteristics.
- Interconnect metal forming properties were examined and potential processes for interconnect structure fabrication evaluated.